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Applications and Advances of Positron Beam Spectroscopy

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Summary

Over 50 scientists from DOE-DP, DOE-ER, the national laboratories, academia and industry attended a workshop held on November 5-7, 1997 at Lawrence Livermore National Laboratory. Workshop participants were charged to address the need for a national center for materials analysis using positron techniques and the capabilities at Lawrence Livermore National Laboratory to serve this need. To demonstrate the need for a national center, the workshop participants discussed both the technical advantages enabled by high positron currents and advanced measurement techniques, the role that these techniques would play in materials analysis, and the demand for the data. Livermore now leads the world in materials analysis capabilities by positrons due to developments in response to demands of stockpile stewardship. There was a detailed discussion of the LLNL capabilities and a tour of the facilities. These facilities now include the world's highest current beam of keV positrons, a scanning pulsed positron microprobe under development capable of three dimensional maps of defect size and concentration, an MeV positron beam for defect analysis of large samples, and electron momentum spectroscopy by positrons.

The workshop included general discussions lead by review talks on positron analysis techniques, and review and focused discussions on applications to problems in semiconductors, polymers and composites, metals and engineering materials, surface analysis and advanced techniques.

The participants concluded that the positron microprobe under development at LLNL and other new instruments relocated to LLNL will take advantage of the world's highest current source of keV positrons will provide an exciting step forward for positron materials analysis. Experiments under consideration for relocation at LLNL include a high resolution electron momentum spectrometer using a focused keV positron beam to perform electronic structure analysis in thin films and at interfaces , a positron re-emission microscope to provide images of single vacancies and other defects, an extremely surface sensitive spectrometer of Auger electrons stimulated by positron annihilation, a high counting rate positron spectrometer for diffraction and holography to provide highly reliable surface structure determinations, and a fully instrumented surface analysis and deposition system for general surface investigations. Each of these experimental techniques was shown to have distinct, significant advantages over existing capabilities based on electrons or photons and the practical application of each of these techniques requires the LLNL positron beam source. New data from these experiments will impact a wide variety of applications. There was unanimous endorsement for establishing a center at LLNL and a plan was devised to build a detailed road map for developing the required technical and fiscal resources.

In discussions of specific application areas, immediate problems were found that require advanced positron techniques and long term issues were identified that will continue and expand those requirements into the foreseeable future.

In analyzing semiconductors there is an immediate need to supply data using a pulsed, scanning microprobe to determine issues of defects and flaws in metallization, defects at the Si/SiO_2 interface and defects in novel materials. In the longer term, positron analysis is expected to contribute to the understanding of the nature of vacancies in Si, the

mechanics of electromigration, processing induced defects, defect complexes and the details of nanostructures.

Polymer and composite analysis is a rich field due to the ability of positron lifetime analysis to determine the size and concentration of open hole volumes in the materials. By performing these analyses, instruments sited at LLNL will enable investigations in new areas of both technical and research interest. Technical interests include a better understanding of the production and evolution of coating-substrate interfaces, stress activated processes of cracking and failure, crazing and degradation of coatings or composite materials, and effects of hardening of polymeric coatings by ion irradiation. Research interests include a clearer understanding of molecular dynamics in polymeric systems and determinations of details of the intrinsic differences in polymer characteristics between the surface and subsurface. Use of the LLNL beams would also allow development of new spectroscopic techniques that would couple electron momentum and electron density techniques to gain a clearer understanding of the details of the positron probe in polymeric systems

Surface analysis techniques of diffraction and Auger emission were shown to have significant advantages when performed with positrons. Positron stimulated Auger emission is significantly more surface sensitive than electron stimulated Auger, and positron diffraction techniques result in more precise determination of the surface structure. Due to the low source strength available at other locations these analyses have only been applied to few systems and parametric studies have not been done. The main barrier to the general application of these techniques will be eliminated at our high current beam.

The high LLNL beam current was also recognized as enabling investigations in new scientific areas. Proposed experiments in positron stimulated desorption will provide a more precise determination of the desorption stimulus than from electrons or photons. Proposed experiments in positron diffraction-holography are only possible with the LLNL beam flux. They will provide superior data to similar experiments performed with electrons. Proposed experiments in annihilation in flight of polarized positrons channeled in single crystals will result in spatial distributions of the electron spins leading to new capabilities in understanding magnetic materials. And relocating the reemission positron microscope to our high current will allow operation at two orders of magnitude higher magnification than presently possible.

Positron analysis of defects and electronic structure in metals, alloys and compounds have been the mainstay of positron materials analysis for two decades. Applying these techniques using LLNL beams and spectrometers to problems in actinides was shown to be complementary to other data obtained by neutron techniques and of interest to specific issues of stockpile maintenance and understanding of actinide metallurgy. Problems for beam analysis at LLNL were also identified in studies of welding, laser hardening and corrosion.

Positron experiments were seen to relate to theoretical advances at several levels. There is a clear role for positron data to validate theoretical models that describe defect production and motion from radiation of strain. These models are at the cutting edge of materials modeling and are central to stockpile stewardship. There are also new demands on theory to provide accurate calculations of observed positron measurements. It was also recognized that the positron can give specific data on one of the most significant outstanding theoretical problems, that of the correlation of the electrons in a solid.

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Introduction

Over 50 scientists from DOE-DP, DOE-ER, the national laboratories, academia and industry attended a workshop held on November 5-7, 1997 at Lawrence Livermore National Laboratory. Workshop participants were charged to address two questions: Is there a need for a national center for materials analysis using positron techniques and can the capabilities at Lawrence Livermore National Laboratory serve this need. To demonstrate the need for a national center, the workshop participants discussed the technical advantages enabled by high positron currents and advanced measurement techniques, the role that these techniques would play in materials analysis and the demand for the data. Livermore now leads the world in materials analysis capabilities by positrons due to developments in response to demands of stockpile stewardship. The Livermore facilities now include the world's highest current beam of keV positrons, a scanning pulsed positron microprobe under development capable of three dimensional maps of defect size and concentration, an MeV positron beam for defect analysis of large samples, and electron momentum spectroscopy by positrons. It was concluded that the positron microprobe under development at LLNL and other new instruments that would be relocated at LLNL at the high current keV source are an exciting step forward in providing results for the positron technique. These new data will impact a wide variety of applications.

General discussion

Positron spectroscopies

Analysis of solids and surfaces can be performed by two kinds of spectroscopy of the positron interaction with the material. Positron annihilation spectroscopies are based on detailed measurements of annihilation gamma rays and will provide details of the electron density or momentum at the positron annihilation site. Examples include positron annihilation lifetime measurements and angular correlation of annihilation radiation. Positron interaction spectroscopies are based on measurement of the positron or associated interaction byproduct. Examples of interaction spectroscopies include diffraction and Auger spectroscopy. Positron interaction spectroscopies often have electron counterparts and are performed in cases when some advantage in sensitivity or analytical discrimination is derived from using positrons.

The positron is the anti-particle of the electron and when introduced into a solid material will quickly slow to thermal energies. After a short period of diffusion the positron will interact with an electron and annihilate producing pure electromagnetic energy, typically in the form of two gamma-rays. Positron annihilation spectroscopies are based on techniques for detecting properties of annihilation gamma-rays and using known relationships to determine properties of the participating electron. Electron momentum is determined by the energy or angular correlation of the annihilation gamma rays and the annihilation rate is determined by the electron density. Thus by collecting the results of spectroscopic measurements of many annihilation events we can determine the solid state electron distributions weighted by the sampling of the positron.

The annihilation site of the positron determines which part of the electron distribution is sampled. Positrons in defect free material span a large volume and sample all of the available electrons. In defect free material, electron momentum spectroscopy can provide detailed information on conduction and valence electron properties such as Fermi surfaces. However in material containing open volume defects the positron will strongly interact and trap at the defect site. The trapped positron is concentrated in the defect and the electron population available for annihilation is confined to the defect volume. The differences between the full electron population and that near the defect volume can be very dramatic. Consequently positron annihilation spectroscopy is a unique, non-destructive tool for the detection of atomic scale defects and measurement of positron annihilation lifetimes can uniquely determine both defect size and concentration.

Spectroscopies of positrons, electrons or ions after positron interaction can provide analytical advantages or unique spatial sensitivity. Positron diffraction data are free of ambiguities found in analysis of electron diffraction. These result from the significantly more complicated angular dependence in the primary electron interactions. Positron induced ionization leading to Auger emission or ion desorption can be performed with very low energy beams providing gentle stimulation and extreme surface sensitivity compared with similar electron techniques. Positron re-emission microscopy can be sensitive to atomic scale defects.

Positron spectroscopies have been used for several decades to investigate selected materials science problems. This success has been achieved in Fermi surface measurements, bulk and depth dependent defect characterization, surface studies by Auger emission and diffraction and other materials analysis using low intensity positron sources. It is generally agreed by interested experimentalists that the lack of access to high intensity positron sources has been a major limitation to the quality and quantity of the data that has been produced by positron spectroscopy . The generally low strength of positron sources is the major limitation to the number of experiments based on positron spectroscopy.

Livermore positron program

Positron spectroscopy has been used for over 20 years at Lawrence Livermore National Laboratory to study defects in metals and compounds and details of the electronic structure of high temperature superconductors and to determine fundamental positron interactions. LLNL capabilities now include the world's highest current keV positron beam at 10^{10} e^+s^{-1}, a 3 MeV positron beam used for beam lifetime measurements and a full complement of experiments performed in the laboratory with commercial radioactive sources, including high resolution electron momentum measurements using angular correlation.

The high current positron beam is produced by techniques pioneered at LLNL and widely duplicated. Low energy positrons are generated by pair production as 100 MeV electrons stop in a tungsten slab. These are then thermalized, accelerated and transported to experimental areas. This technique is the only demonstrated technology capable of reliably producing high current positron beams. The beam can be used with the time distribution determined by the parameters of the linac electron beam or stretched to produce a quasi-continuous beam of positrons for future experiments. A switchyard has been placed at the end of the stretcher to route the beam to several experimental stations.

LLNL is currently developing a pulsed positron microprobe that will modify the quasi-continuous beam to produce a pulsed, focused positron beam to perform positron beam lifetime experiments with a one micron beam. By varying the energy and lateral position of the pulsed, focused beam, 3-dimensional scans of defect size and concentration will be possible on a practical time scale with sub-micron resolution.

The 3 MeV positron beam is produced by capturing positrons from a ^{22}Na radioactive source installed in the high voltage terminal of a Pelletron electrostatic accelerator. The high energy beam lifetime spectrometer operates with a 50 mCi ^{22}Na source to provide a current of 6 10^5 positrons per second. Lifetime data are determined from a thin plastic transmission detector providing an implantation time and a BaF_2 detector to determine the annihilation time. Positron lifetime analysis is performed with a 3 MeV positron beam on thick sample specimens at counting rates in excess of 2000 per second. The instrument is being used for bulk sample analysis and analysis of samples encapsulated in controlled environments for in situ measurements.

All of the elements of the LLNL positron program are located on the LLNL site at building 194 and are readily accessible to experimenters world wide. The facilities have sufficient space and technical capability to host additional experiments that would benefit from the high beam current.

Applications of positron spectroscopy

Applications of positron spectroscopy were discussed in review and focused sessions. Detailed discussions of the focus sessions are presented below by the session leaders and all of the transparencies from the workshop can be found in Applications and Advances of Positron Beam Spectroscopy: Appendix A CONF-9711105, which is available from the University of California.

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Reports from focus sessions:

Applications of the positron technique to problems of current materials science were discussed in detail. The following are summaries of each of the focus session discussions as reported by the session leaders.

Semiconductors

Session leader: John M. Poate

The studies of semiconductor materials and structural properties are crucial to the advancement of the subject. While analytical techniques such as transmission electron microscopy, secondary ion mass spectroscopy and Rutherford backscattering have proven to be essential workhorses in these studies, the case still has to be made for the rôle of positron beam spectroscopy. In this report, members of the semiconductor focus group have identified research areas where the identification and analysis of vacancy or void - like defects is of importance to semiconductor science and technology. Most of the areas focus on Si and deal not only with the semiconductor itself but also the overlying metallization schemes.

Low Hanging Fruit or Immediate Studies

The following areas are those where the identification of free volume defects could have immediate and important ramifications

Metallization Studies -
The multi-level metallization schemes pose the greatest materials challenge in Si technology. Positron beams could have a unique niche for determining voids in vias and properties of low k dielectrics.

Si/SiO$_2$ Interfaces Studies -
Previous positron studies have postulated the existence of voids at the oxide/semiconductor interface. This finding is of such potential importance that it should be confirmed as soon as possible.

Novel Materials Analysis -
Novel materials are being investigated, for example, for gate oxide replacement or low k dielectrics for metallization. Positron beams could prove important probes of these materials.

Core or Longer Range Studies

Nature of Vacancies in Si -
Positron spectroscopy played an important role in the elucidation of vacancy behavior in metals. Equivalent efforts need to be devoted to Si.

Mechanisms of Electromigration -
Much remains to be understood regarding the generation of vacancies by the electron wind in high current density conductor stripes. This is an obvious area for a major positron effort.

Process Induced Defects -
High energy implantation causes the separation of interstitials and vacancies, and positron spectroscopy is the best tool for identifying the vacancies; these defect structures have to be evaluated as high energy implantation is now being widely implemented. Other processing areas, such as silicide formation, should be investigated for vacancy formation.

Complex Defects -
Point defect structures in compound semiconductors such as GaN tend to be complex in nature and the role of vacancies should be elucidated.

Nano structures -
This rapidly growing field, dealing with such subjects as quantum dots, quantum wires and self assembly of small clusters, is ready for defect analysis by positron spectroscopy.

Polymers and Composites

Session leader: J. Jean

Eight people presented papers at the Polymer Focus Section on the workshop. Dr. P. Wu is from Boeing Aircraft Company, and the rest are university scientist who have been working in positron research. Their presentations are summarized in terms of the three following key questions:

Scientific issues in polymers

Applications of positron spectroscopy to polymeric materials have been particularly successful in recent years.[1] Due to the localization of positronium (Ps) in subnanometer defects such as holes, voids and free volumes, positron spectroscopy has emerged as the only method to provide both qualitative and quantitative information. Clear correlations between the positron parameters (o-Ps lifetime, intensity, and S parameter) and materials properties, glass transitions, gas diffusion, and local volume expansivity have been established. The use of positron spectroscopy has been extended to search for engineering applications of polymers, such as modulus, toughness, environmental protections, and processing.

The most important issue is the ability of positron spectroscopy to detect problems at the initial stage of degradation in polymeric materials. Existing probes, such as TEM, AFM, and X-rays, detect degradation in its final stage. In this respect positron spectroscopy can be used to prevent, or provide clues to solve degradation problems (such as weathering, physical aging, creeping, fatigue, cracking, etc.) of polymeric materials under various uses.

The other important scientific issues is the imaging of three-dimensional defect structures for microvoids. Creep and crazing of polymeric materials are crucial problems in engineering applications. Microvoid sizes commence at the μm level, which can be probed only by a microprobe. The subnanometer defect size around these microvoids will be mapped as a function of depth from the surfaces. This will provide key information about the formulation of theses types of microvoids at the early stage.

The need for advanced techniques

While the majority of positron studies in polymers have been for the bulk state, a few existing surface studies have shown that high sensitivity and specialization are needed to probe the surface and interfacial problems. [2,3] Two advanced techniques are:

(1) Spatial Resolution (sub-microns):

As mentioned in the above section on the important scientific issues, advanced microbeam techniques will open a new area of scientific understanding in the problems of microvoid structures from the surface down to the bulk.

(2) Time Advantage with Intense Flux (10^9 e^+/sec):

The most important property of polymeric materials is their dynamic behavior. Their visco-elastic properties are time-dependent. Advanced microbeam technique provides an intensity high enough to acquire a positron datum in a much shorter time, such as seconds. This will open a new area of monitoring the propagation of degradation mechanism in polymeric materials

Special requirements

In polymeric applications, high resolution ($\leq$ 250 ps) positron lifetime annihilation spectroscopy is essential. A high counting rate (10^6 s^{-1}) will enable us to search for a shorter time interval (subsecond) of dynamic behavior. A smaller beam size (submicron) will also provide better spatial resolution to map the defect structure. A positron energy of ~ 10^6 eV will be required to investigate deep bulk properties. In addition to positron annihilation lifetime spectroscopy, the following state-of-the-art positron annihilation instruments are required for complete information: two dimensional angular correlation of annihilation measurements of electron momenta, simultaneous electron momenta-electron density measurements, positronium velocity measurements by time of flight, and Doppler broadening spectrometers of electron momentum distributions.

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2. G.B. DeMaggio, W.E. Frieze, D.W. Gidley, M. Shu, H.A. Hristov, and A.F. Yee, *Phys Rev. Lett.* 78, 1524 (1997).
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Theory of Positron Annihilation in Condensed Matter

Session leader: Warren Pickett

The spectroscopy of condensed systems arises from probing them with elementary particles: photons, electrons, neutrons, positrons, muons, or ions. There are only a few such elementary particles, so each is important and each is unique in its own way. The particles interact with the constituents of the solid, which are electrons and nuclei. Understanding of the behavior of the electronic system is the central problem of condensed matter science. The positron is special in that it has the mass and magnitude of charge of the electron, and thus reflects quantum mechanical effects in a closely related way. Indeed the study of positron annihilation in condensed systems is a mature science and a tremendous amount has been learned about the properties of matter from positron studies.

The fundamental feature of positron probes is that annihilation arises from the interaction of the positron with the electron - it probes in detail the electron-positron wavefunction overlap and gives very specific information about the electron wavefunctions. Because the positron is repelled from the nuclei, it samples the *valence* electron states preferentially. This bias is almost always desirable, but it has a crucial implication: because the valence electrons are environment-sensitive, it is essential to calculate self-consistent, quantum-mechanically correct wavefunctions for the electrons as well as for the positrons.

Fortunately, the computational theory for calculating both electronic and positronic wavefunctions is well in hand in many respects. A spectacular example is that of the high temperature superconductor $YBa_2Cu_3O_7$ T_c=93 K. The multi-sheeted Fermi surface of this important compound was calculated well before it was confirmed by experiment. Applications in determining Fermi surface dimensions and characters remains a very important aspect of positron spectroscopy, and a crucial one in the case of short mean free path materials. An example is the very recently reported observation of the Fermi surface in the correlated metal compound $CeCu_2Si_2$.

The sensitivity to valence electrons makes positrons a uniquely suitable probe of electron correlation. The fundamental physics of electron correlation effects plays an important role in transition metal oxides, intermediate valence materials, high temperature superconductors, heavy fermion superconductors and Kondo systems as well as the elemental actinide metals. Positrons probe the momentum associated with the occupied electron states, including all correlation effects, so measurements of the momentum tails associated with the f-electron states can yield important information about the nature of electron correlation and localization-delocalization transitions. The combination of positron 2D-ACAR and Compton profile data on alkali metals can provide a test of the conventional self-energy-based models for electron correlation by measuring the momentum distribution at values larger than the Fermi momentum. Theories of electron correlation focus primarily on the energy dependence of the electron states in the vicinity of the Fermi surface due to the availability of experimental techniques like photoemission that probe this variation. More theoretical work is needed on the momentum distribution of electrons in correlated systems, where positron annihilation may provide new insight into this important branch of condensed matter physics. A high intensity positron source, advanced materials growth and characterization techniques, and appropriate theory are all required to make significant progress in this area.

Fermi Surfaces. The Fermi surface, where the low energy excitations of a metal live, is the most fundamental property of a metal. Two dimensional angular correlation of annihilation radiation (2D ACAR) measures the momentum density of the interacting electron-positron system, which has discontinuities at the Fermi surface. Identification of the Fermi surface of a material requires a momentum density calculation, because even in very favorable cases the momentum density can change substantially within the resolution of the data, and because poor overlap of the positron with some electrons on the Fermi surface can render the discontinuities tiny. In calculating the positron distribution, it is necessary to know the electron-positron correlation functional. For most purposes the current knowledge of this functional is quite sufficient. In some delicate situations it is a close call in which part of the cell the positron is located, or whether the positron is trapped or not. For such cases improvement of the functional will be required.

Electron Momentum Measurements of Defect Sites. Momentum measurements and calculations enable the identification of chemical species in the vicinity of a vacancy as well as permitting the identification of electronic structure features such as Fermi surfaces. Defects in the vicinity of interfaces can be studied using variable energy positron beams. The accuracy of the calculated positron distribution in the defect is again a significant issue, and more joint experimental-theoretical work is needed to demonstrate the general applicability, reliability, and potential importance of these techniques.

Positron Lifetimes. Calculated positron lifetimes provide a basis for interpreting experimental data by providing the expected annihilation rates for bulk and defect states. There is very good agreement between experimental results and first principles theory. However, the clear comparisons are limited to well-characterized simple systems such as elemental materials and simple intermetallic compounds. Experience on more complicated systems is very limited, although encouraging results have been obtained (e.g. BKBO). Nevertheless, considerably more data on well characterized systems is required before we can say with confidence that the theoretical calculations are reliable for a wide range of materials. The positron facility at LLNL will allow a significant increase in the number of such experiments, and thereby result in a broader set of experimental data for validating the theoretical calculations.

Theory and Computation. There are some clear theoretical needs in the general positron community, and some specific theoretical and computational needs within the context of the LLNL positron facility. First, data analysis tools are required for extracting the maximum amount of information from the experimental data. These tools should be sufficiently easy to use so that non-positron experts can obtain useful information from their experiments at the facility. Second, first-principles calculations of lifetimes or momentum spectra are needed to guide the interpretation of complicated experimental data. Third, a wide range of fundamental positron science issues remain unresolved, or are treated in only an approximate way in current approaches. For example, improvements in the description of the electron-positron correlation energy and the enhancement of electron - positron overlap by many body effects will improve the quality of these calculations. Better descriptions of positron implantation and diffusion, including lateral dispersion, will be essential for interpretation of data from the positron microprobe. Backscattering and transmission experiments can provide a basis for validation of existing implantation models based on Monte Carlo simulations, and indicate areas where improvement is called for. Additional effects, such as positronium formation in polymers and at surfaces and the trapping of slow positrons in surface states, are largely unexplained. A better theory of these phenomena is required to understand positron free-volume measurements in polymers as well as the efficiency of positron remoderators. Finally, there are a class of experiments that positrons can perform to provide parameters or guidance for the construction of models for materials behavior. Measurements of vacancy formation energies and diffusion fit in this category, and represent an important area where theorists may become clients of the positron facility.

Since the interpretation of positron annihilation experiments requires sophisticated theoretical calculations, the plan for the positron facility should include a theoretical component to complement the planned experimental facilities. This theoretical component should include computational facilities for data analysis and theoretical calculation of spectra with associated funding for on-site staff as well as funding for off-site research on theoretical issues that are central to the unique experimental techniques available at the facility.

REFERENCES

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Applications of Intense Positron Beam for Surface Materials Characterization

Session leader: S.Y. Tong

The versatile properties of the antimatter particle-the positron-are utilized in unique experiments to probe materials properties in the surface region. The experiments and accompanying analyses are often superior in accuracy, sensitivity or resolution compared to measurements involving electrons, photons or ions. We mention below four experiments which utilize various properties of the intense positron beam:

Surface Diffraction and Holography for Materials Characterization, including Biological Materials

These experiments utilize the elastically scattered positrons by matter. In surface diffraction, scientist have long searched for an ideal probe to study the structure of matter. The particle of the ideal probe must satisfy two criteria. First, the particle must have a shallow penetration in a solid to ensure its surface sensitivity. Second, the particle must weakly interact, i.e. scatter, with the atoms in solid. A weak scattering interaction results in diffraction spectra which are simple to analyze.

A positron beam has attributes which satisfy the above two criteria. A positron has an identical mass as the electron; however, it has a positive charge while an electron has a negative charge. A positron has a shallow penetration distance in a solid because this distance is determined by plasma generation in a solid. The inelastic process is independent of the sign of the probe particle. Therefore, a positron satisfies criterion No. 1. In addition, a low-energy positron scatters weakly inside a solid because the positive charge of the positron and that of the atomic ion cores repel each other. Therefore, a positron can never closely approach the core region of an atom for it to be strongly scattered (1).

Recent works have shown that the weak scattering of positrons in solids allows a more accurate interpretation of its diffraction spectra by simplified scattering methods. In addition, direct methods such as holographic wavefront reconstruction work better for inverting positron diffraction spectra than for electron diffraction spectra (2). The latter is due to the factor that positrons are not bound by atomic potentials and the positron's scattering factor does not contain resonance lobes which involve jumps of near pi in the scattering phase. Because of the weak scattering of positrons by atoms, the structure of large biological molecules can be studied by coherent beam positron diffraction.

Positron Stimulated Auger Emission Spectroscopy

Positron Annihilation Induced Auger Electron Spectroscopy (PAES) makes use of a beam of low energy positrons to excite Auger transitions by annihilating core electrons. This novel mechanism gives PAES a number of unique capabilities as a surface analytical tool. In PAES the very large collisionally induced secondary electron background which is present under the low energy Auger peaks using conventional techniques can be eliminated by using a positron beam whose energy is below the range of Auger electron energies. In conventional photon or electron excited Auger spectroscopy, the secondary electron background in the Auger electron energy range cannot be avoided since the incident beam energy must be greater than the core level ionization energy (and hence greater than the Auger electron energies). Another advantage provided by positrons is that of extreme surface selectivity: the PAES signal originates almost exclusively from the topmost atomic layer due to the fact that the positrons annihilating with the core electrons are trapped in an image correlation well just outside the surface. Results of experiments on clean and adsorbate covered surfaces demonstrate the extremely low background, and increased surface sensitivity that is possible with PAES. PAES provides advantages for the study of growth, alloying and inter-diffusion of ultrathin layers of metals on metals, metals on semiconductors, and semiconductors on semiconductors.

Positron Induced Desorption of Ions

With the realization of the large-flux dc or time-bunched positron source(10^{10} e^+/sec) recently commissioned at LLNL, electronic desorption experiments previously only dreamed of are now within our reach. This unique capability allows us to study fundamental processes leading to desorption at well characterized surfaces in totally new ways. These studies are intrinsically important to an understanding of desorption which in turn leads to a much enhanced understanding of growth phenomena. Large flux beams of positrons allow us to create well-defined initial states due to core-hole formation by matter/anti-matter annihilation (not by collisional ionization) at well defined localized sites at the surface and in the near surface bulk. Electronically induced desorption processes at surfaces may come about both by direct bond breaking processes as well as by indirect defect moderated mechanisms. By varying the energy of the positron beam, hole formation may be accomplished exclusively at the surface or at a well defined deposition depth below the surface. By this means, we will be able to identify particular defect dependent processes leading to surface desorption and to measure defect migration lifetimes. This will be particularly applicable to optical materials and thin film semiconductors. The following is a partial list of unique experimental opportunities now made possible by this source:

* Probe the microscopic defect mediated process leading to surface desorption, and determine defect migration lifetimes as a function of experimental parameters (i.e. temperature and depth dependence).

* By using a coincidence technique one may correlate the desorbed particle with the specific core hole energy level and momentum and obtain cross-section for various electron energy levels.

* One can monitor the transition from collisionally induced desorption to the energy regime for which only annihilation processes occur.

* Using the time resolved capabilities of the positron beam one can measure the kinetic energy of the desorbed ion and estimate the excited state of the adsorbed species in the time range of tens to thousands of picoseconds.

Desorption studies have in the past been hampered by the virtual impossibility of creating well defined initial states. The recent availability of the large flux positron source provides an unparalleled opportunity to elucidate basic dynamic processes leading greatly enhanced understanding of desorption and growth phenomena.

Positron Emission Microscopy

The positron re-emission microscope images positrons emitted from materials having a negative positron workfunction. A positron beam is focused to a small spot on a thin foil and a magnified image of the emitted positrons is formed after diffusion through the foil and expulsion into the vacuum. Since the diffusing positron can be trapped by vacancies and other defects one can directly image sub-surface structures that lack sufficient contrast in existing electron microscopes. A second generation microscope ultimately capable of 1 nm resolution is currently in operation with a 100 mCi radioactive source. Resolution in this mode is limited to 10 nm and dislocations have been imaged in Ni (100) films. Establishing this instrument at an intense beam will allow experiments to image the spatial distribution of a set of mono vacancies at a buried interface, to image mis-fit dislocations, to image details of surviving vacancies from ionizing radiation tracks and to image interfacial defects in heteroepitaxial island growth.

Special requirements

For the above experiments, surface diffraction and holography and positron stimulated Auger spectroscopy can be carried out in modified general purpose surface science chambers. The experimental chambers for experiments on positron stimulated ion desorption and positron reemission microscopy already exist. All of these experiments require a beam as intense as found at LLNL. Some additional site preparation may be required to match the designs of the existing chambers to the LLNL beam.

(1) S.Y. Tong, H. Huang and X.Q. Guo, PHYS.REV.LETT, 69 3654 (1992).

(2) "Physics in Action-Positron Challenge Electrons in Surface Studies". Physics World 6, No. 3 (1993).

Engineering and Engineered Materials

Session leader: Roy West

Studies of metals and alloys have been a strong and persistent theme throughout the history of slow positron physics and, since the discovery of the positron-defect trapping phenomena in the late sixties and the subsequent numerous successful applications of that phenomenon to a wide range of metallurgical problems, they have provided the best examples of genuine materials science. The positron is a uniquely sensitive probe of open-volume defects and of electronic structure but, in both of these areas, the inadequate brightness of the radioisotope or low-intensity laboratory beam sources has seriously limited the realization of its full potential. It has produced startling pictures of the Fermi surface of electronically complex conductors, a catalogue of vacancy formation enthalpies and other valuable data but only where we have had, or could synthesize, samples of a sufficient volume to stop the incoming positrons and a homogeneity sufficient to make the (necessarily volume averaged) data relevant to the equilibrium thermodynamic quantities involved. ACAR measurements of the electronic structure of high temperature superconductors and other complex compounds have been made more difficult or sometimes impossible by the high concentrations of intrinsic defects in them and the modest quality of crystals of a sufficient size for study. If, in a metallurgical study, the system or the process of interest was heterogeneous, the sensitivity of the measurement was reduced and the interpretation of the results could, at best, only be qualitative.

The problems occasioned by volume averaging have been reduced with beam positrons where depth resolution obtains, but only marginally so. However, the overall flux of the Livermore beam, its continuous and high repetition rate pulse modes of operation and its microprobe station will overcome these and other limitations of the current sources. We would note the following possibilities.

1. The high flux continuous mode of operation will enable precision ACAR studies of electronically complex compounds in micro-crystals of much better quality than the large crystals thus far studied, on thin films and in multi-layers. At a still more fundamental level, studies of fabricated low-dimensional systems, such as arrays of quantum structures, are an exciting possibility.

2. The microprobe scanning facility will provide full three-dimensional (lateral and depth) spatial resolution and will, for the first time, make possible studies of defects in small sampling volumes. An associated number of applications come immediately to mind in areas such as coatings (delamination), corrosion (impurity driven and stress induced) embrittlement and fracture. All these phenomena arise from heterogeneous processes and positron implantation into small volumes should result in sufficient sensitivity for investigation of the earliest stages and, in some cases, real-time monitoring of a developing situation. ORNL is interested in examining states resulting from weld-pool solidification and this and other non-destructive evaluation applications seem likely.

3 In all modes of operation, lifetime, Doppler broadening and combination techniques such as age-momentum and coincidence Doppler measurements will obviate the current source-sampling packaging problem and allow studies of vacancy formation and migration and other basic parameters in a wider range of toxic, reactive and refractory materials.

Advanced Techniques

Session leader: K. G. Lynn

In an attempt to look to the future for the LLNL positron beam one needs to consider the new techniques or instruments that will be needed to fully utilize the high flux and brightness capabilities of the proposed facility. This new positron system promises to provide images of a wide variety of subjects with entirely new types of contrast and measurements with new instruments that will provide qualitatively improved structure determinations for various solids and surfaces of importance to science and industry. A well equipped positron facility would be a dream come true for experimenters who have struggled for years with small positron sources. Hundreds of researchers would benefit from an easily used set of instruments at this intense positron source. Demand for a facility has accumulated for almost 20 years, since the first attempts at BNL, and various unrealized plans at INEEL, Takashashi and other institutions. The positron experimenters of the US and beyond will welcome this facility.

Some of the goals of an intense beam positron facility include obtaining important information such as::

- Images of voids in the metallization of an integrated circuit chip.
- Positron contrast views of tobacco mosaic and other viruses
- Reconstruction of single protein molecules seen by speckle diffraction
- Images of the electronic bands in high Tc superconductors
- Positron induced Auger images of the chemistry of crack tips
- Three- dimensional mapping of the electronic structure and defects using the positron lifetime and momentum of various systems
- Time resolved behavior of transient stressed solids
- Real time images of the interactions of individual vacancies and dislocations with impurities in materials under actual technologically important conditions.
- Ultrahigh resolution in angle and energy measurements of the electronic band structure of metals
- Images in real time of the interactions of individual vacancies in materials at low temperatures.

This list does not include the variety of fundamental measurements that will be possible with this new capability. Both the applied and fundamental problems require state of the art instruments which are presently unavailable at any facility. The instruments and enhancements required include:

State of the Art Angular Correlation Apparatus –
To fully utilize the flux of this facility, an enhanced angular-correlation apparatus is needed. This instrument requires high spatial resolution (1-2 mm), efficiency and the incorporation of a large solid angle. Such an angle can be accomplished using multiple sets of Anger cameras, or a series of two dimensional photomultiplier tubes coupled with bismuth germanate scintillation crystals. Both of these systems exist in the commercial sector. Cost and the exact beam characteristics will determine which system is used.

Positron Holography System -
This system could be constructed from commercially available sources. The vacuum system will be constructed from a fully developed system being donated from Canada.

Speckle Diffractometer System –
A highly efficient positronium velocity spectrometer is required to measure the electronic surface structure of thin films.

Positron Stimulated Desorption System coupled with the Positron Induced Electron Chamber.
Desorption studies have in the past been hampered by the virtual impossibility of creating well defined initial states. The recent availability of the large flux positron source provides an unparalleled opportunity to elucidate basic dynamic processes leading greatly enhanced understanding of desorption and growth phenomena.

Improvements to the existing positron re-emission microscope -
Further development of the positron microprobe beam presently under construction is necessary to perform real time experiments under unique experimental conditions. Such

conditions require high and low temperature capabilities, a stress and strain apparatus, and a separate load lock chamber to carry out anneals in a variety of high pressure and temperature experiments.

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